BEFORE THE HON'BLE NATIONAL GREEN TRIBUNAL SOUTHERN BENCH

AT CHENNAI

O.A. 246 OF 2024

In the matter of News item title "Zinc, Lead…fish in Kochi estuary are heavy, daily intake risky" appeared in The Times of India dated 06.05.2024

…Petitioner

Vs.

National Centre for Coastal Research, through its Director,
Chennai & Ors., …Respondents

INDEX

DATED AT CHENNAI ON THIS THE 4TH DAY OF JANUARY 2025

COUNSEL FOR 1ST RESPONDENT

1.

BEFORE THE HON'BLE NATIONAL GREEN TRIBUNAL (SOUTHERN BENCH) CHENNAI

O..A. 246 OF 2024

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REPLY AFFIDAVIT OF NCCR

I, K. Venkatarama Sharma, Son of Late V. Sankara Krishna Sharma, aged about 58 years, occupying the Post of Scientist F in the National Centre for Costal Research having their office at NIOT Campus, Velachery-Tambaram Road, Pallikaranai - 600100, do hereby solemnly affirm and sincerely state as follows:-

1. I am occupying the post of Scientist F in the National Centre for Coastal Research and I am well acquainted with the facts of the case.

2. I submit that this Hon'ble Tribunal has taken a suo moto cognizance of the scientific findings, published in The Times of India, a daily News Paper, highlighting heavy metal pollution in the Kochi estuary including the Periyar River in Kerala. The news also appeared in both electronic and print media in Kerala. I submit that the reports in these daily journals were based on the collaborative work carried out by Cochin University of Science and Technology (CUSAT) and the National Centre for Coastal Research (NCCR), MoES, Government of India. The scientific outcomes of the project were published in the peer-reviewed reputed Springer- Journal, Toxicology and Environmental Health Sciences (Published on 30 April 2024; Vol.16; pp. 217-231; ISSN 2233-7784 – **Annexure I**), after a thorough review process by experts in the relevant field. The outcome of the study is part of a national Grant-in-Aid network project implemented at the Department of Marine Biology, Microbiology, and Biochemistry at the School of Marine Sciences, Cochin University of Science & Technology (CUSAT) and funded by the

के. वेंकटरामा शर्मा
K. VENKATARAMA SHARMA
वैज्ञानिक-एफ / Scientist - F
राष्ट्रीय तटीय अनुसंधान केन्द्र
National Centre For Coastal Research
पृथ्वी विज्ञान मंत्रालय
Ministry of Earth Sciences
भारत सरकार, एन आई ओ टी परिसर
Govt. of India, NIOT Campus
पल्लिकरणै, चेन्नै-६०० १००.
Pallikaranai, Chennai - 600 100.

Ministry of Earth Sciences * NCCR Govt. of India *

National Centre for Coastal Research (NCCR), Ministry of Earth Sciences, Government of India, Chennai.

3. In respect of the concerns raised by this Hon'ble Tribunal in Para 1 to 4 of the Order dated 02.07.2024, we submit that the study establishes that Kochi coastal waters is facing environmental issues, primarily due to industrial activities, waste disposal from developmental projects, sewage, fishing, tourism, and other non-point source of pollution and it is difficult to establish the source. The study locations selected for the current study are associated with the Periyar river system, near to the industrial zone where recurring "fish kill" from anoxia and heavy metal contamination and related issues are frequently reported.

4. The field-based samples of water, sediment, fish and shell fishes were collected during 2021 from the Southern (estuarine zone), Northern (riverine, including Periyar river zone) and Central zone (marine) of Kochi coastal waters; compared to studies from the past decade, metal concentrations in the coastal waters are increasing, indicating a rise in pollution load, particularly in the Kochi environs. The metals Cu, Zn, Pb, Cd, Ni, and Cr were found at increased concentrations in the northern zone of the estuary. Similarly, sediment samples also showed the highest concentrations of metals (Cu, Zn, Mn, Ni, and Cr) in the northern estuary. The geo-accumulation index indicates that Cd contamination was moderate in the central and northern parts of the estuary. Results indicated that Zn, Cd, and Cr levels in the sediment exceeded the Probable Effect Limit (PEL) and Effect Range Median (ERM) set by National Oceanic and Atmospheric Administration standards (NOAA, 1999), implying significant adverse effects on fishery and other resources.

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5. The evaluation of potential human health risks from exposure to heavy metals depends on the target hazard quotient (THQ) and hazard index (HI). The THQ assess the health risks of individual metals and HI evaluates their synergistic effects. If the values of THQ and HI exceed 1, there is a possibility for non-carcinogenic health risks. In this investigation, Cd showed the highest THQ (0.25 ± 0.08), ranging from 0.12 (*P. monodon*) to 0.37 (*Thryssa mystax*), while Mn exhibited the lowest THQ ($8.82 \times 10^{-3} \pm 6.76 \times 10^{-3}$), with a range of 2.9×10^{-3} (*T. mystax*) to 2×10^{-1} (*C. ignobilis*). The THQ values for the metals followed the order: Cd > Cr > Cu > Pb > Ni > Zn > Mn. *The 95th percentile value of the HI was 0.80, suggesting that there is no significant risk of non-carcinogenic health effects from consuming these fish and shell fishes.*

6. The carcinogenic risk is negligible, if the target cancer risk (TCR) index is $\leq 10^{-6}$, unacceptable if the values are $>10^{-4}$ and acceptable if the values are between 10^{-6} and 10^{-4}. The 95th percentile TCR for Pb, Cd, and Cr were 6.54×10^{-6}, 2.15×10^{-3}, and 3.90×10^{-4} respectively. It indicates a possibility of cancer risk from Cd. The sensitivity analysis indicated that the non-carcinogenic and carcinogenic risks are primarily affected by metal concentration. *Overall, the study inferred that the possibility of non-carcinogenic risks, such as liver and kidney disorders, respiratory issues, dermal toxicity, etc., from consuming these seafood resources are minimal.*

7. I submit that the study had also recorded that estimated daily intake (EDI) of these metals are well within the suggested daily intake (RfD) and there is no significant risk of non-carcinogenic health effects by the metal accumulation in fishes and shell fishes based on target hazard quotient (THQ) and Pb, Cd, and Cr may cause possible cancer risk. However, the cancer risk by metals should be confirmed with further studies.

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Ministry of Earth Sciences NCCR Govt. of India

8. I submit that in so far as the concerns raised by this Hon'ble Tribunal in Para 5 of the Order dated 02.07.2024, I submit that the metal concentrations in the ambient waters are regulated by enforcing the Primary Water Quality Standards under the Environment Protection Act, of 1986. These water quality standards are notified for compliance in ambient water for the designated best-use classes I-V. Presently, the primary water quality standards for coastal waters are available for three metals only based on expert opinion **(Annexure II).** The standards for other metals are being perused by CPCB of MoEF&CC, where NCCR extends the scientific support.

9. I submit that the Comparison of the values with the primary water quality standards with additional seven metals (Cd, Cu, Cr, As, Pb, Hg, Zn) and Pesticide was developed by NCCR and published on MoEF&CC website, the values are higher for Zn and Pb while it is within the range for Cu, Cd, Cr. Given the above, this Respondent fully agrees with the observation of this Honourable Tribunal that standards for these concerned metals also need to be complied with as per the implementation and enforcement by MoEF&CC and CPCB through coastal state SPCBs and PCCs of UTs to ensure the ambient water quality. Therefore, the regulatory standards of coastal environmental contaminants should be achieved through preparing National Action Plan by State Government by engaging expert national institution.

10. As part of notification of amendment in primary water quality standards, compliance of primary water quality standards in marine outfalls across Indian coastal waters has been investigated by NCCR in collaboration with coastal state SPCBs and PCCs of UTs. The marine outfall compliance report was submitted to MoEF&CC and CPCB. Mostly the concentrations of metals (Cd, Cu, Cr, Hg, Pb, As and Zn) in marine outfalls are within the standards with few exceedances.

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11. Therefore, in view of the above it is suggested that this Hon'ble Tribunal may instruct state government to prepare National Action Plan for abatement of pollution and to improve the health of coastal waters.

(K. VENKATARAMA SHARMA)

Solemnly affirmed at Chennai on this the 30th day of August 2024 and signed his name in my presence after reading the contents herein

* BEFORE ME,
*
* ADVOCATE, CHENNAI

Ms/2577/23
No-366, Law Chambers, Highcourt building, ch-104

के. वेंकटरामा शर्मा
K. VENKATARAMA SHARMA
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राष्ट्रीय तटीय अनुसंधान केन्द्र
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…Respondents

COUNTER

S. JANARTHANAM(877/91)
COUNSEL FOR 1ST RESPONDENT

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रजिस्ट्री सं. डी.एल.- 33004/99 REGD. NO. D. L.-33004/99

भारत का राजपत्र
The Gazette of India

सी.जी.-डी.एल.-अ.-18022020-216258
CG-DL-E-18022020-216258

असाधारण
EXTRAORDINARY

भाग II—खण्ड 3—उप-खण्ड (i)
PART II—Section 3—Sub-section (i)

प्राधिकार से प्रकाशित
PUBLISHED BY AUTHORITY

सं. 104] नई दिल्ली, सोमवार, फरवरी 17, 2020/माघ 28, 1941
No. 104] NEW DELHI, MONDAY, FEBRUARY 17, 2020/MAGHA 28, 1941

पर्यावरण, वन और जलवायु परिवर्तन मंत्रालय

अधिसूचना

नई दिल्ली, 17 फरवरी, 2020

सा.का.नि. 122(अ).—अधिसूचना, जिसे केन्द्रीय सरकार, पर्यावरण (संरक्षण) अधिनियम, 1986 (1986 का 29) की धारा 6 और धारा 25 में प्रदत्त शक्तियों का प्रयोग करते हुए जारी करने का प्रस्ताव करती है, का निम्नलिखित प्रारूप पर्यावरण (संरक्षण) नियम, 1986 के नियम 5 के उपनियम (3) की अपेक्षानुसार, जनसाधारण, जिनके उसके द्वारा प्रभावित होने की संभावना है, की जानकारी के लिए, एतद्द्वारा प्रकाशित किया जाता है; और एतद्द्वारा सूचना दी जाती है कि उक्त प्रारूप अधिसूचना पर उस तारीख से, जब से भारत के राजपत्र की प्रतियां, जिसमें यह अधिसूचना अंतर्विष्ट है, जनसाधारण को उपलब्ध करा दी जाती हैं, साठ दिन की अवधि की समाप्ति पर या उसके पश्चात विचार किया जाएगा।

ऐसा कोई व्यक्ति, जो प्रारूप अधिसूचना में अंतर्विष्ट प्रस्तावों पर कोई आपत्ति या सुझाव देने में हितबद्ध है, इस प्रकार ऊपर विनिर्दिष्ट की गई अवधि के भीतर, केन्द्रीय सरकार द्वारा विचार किए जाने के लिए, आपत्ति या सुझाव सचिव, पर्यावरण, वन और जलवायु परिवर्तन मंत्रालय, इंदिरा पर्यावरण भवन, जोर बाग रोड, नई दिल्ली - 110003 को लिखित रूप में या ई-मेल पते अर्थात् mscb.cpcb@nic.in और h.kharkwal@nic.in पर सदस्य सचिव, केन्द्रीय प्रदूषण नियंत्रण बोर्ड और मंत्रालय के वैज्ञानिक 'ई' को भेज सकेगा।

प्रारूप अधिसूचना

केन्द्रीय सरकार, पर्यावरण (संरक्षण) अधिनियम, 1986 में और अधिक संशोधन करने के लिए एतद्द्वारा निम्नलिखित नियम बनाती है, अर्थात् -

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1. संक्षिप्त शीर्षक और प्रारम्भ.—(1) इन नियमों को पर्यावरण (संरक्षण) संशोधन नियम, 2020 कहा जाएगा।

(2) ये आधिकारिक राजपत्र में उनके अंतिम प्रकाशन की तारीख से लागू होंगे।

2. पर्यावरण (संरक्षण) अधिनियम, 1986 में, अनुसूची-I में तटीय जलक्षेत्र समुद्री निकास के लिए जल गुणवत्ता मानकों से संबंधित क्रम संख्या 86 के लिए निम्नलिखित प्रविष्टियां प्रतिस्थापित की जाएंगी अर्थात्:—

(क) तालिका 1.3-श्रेणी एसडब्ल्यू-III जलक्षेत्र [औद्योगिक, शीतलन, रीक्रिएशन (गैर-संपर्क) और सौंदर्यशास्त्र] के लिए प्राथमिक जल गुणवत्ता मानदंड के नीचे, क्रम संख्या 9 से 16 पर, क्रम सं. 8 के बाद अतिरिक्त मापदंड निम्नानुसार प्रतिस्थापित किए जाएंगें:—

क्र. सं.	मापदंड	मानक	औचित्य/अभ्युक्ति
9.	कैडमियम (सीडी के रूप में)	3.03 माइक्रोग्राम/लीटर या कम	मानदंड संबंधी सतत सांद्रण (सीसीसी) के आधार पर **
10.	तांबा (सीयू के रूप में)	4.1 माइक्रोग्राम/लीटर या कम	
11.	पारा (एचजी के रूप में)	0.38 माइक्रोग्राम/लीटर या कम	
12.	जस्ता (ज़ेडसी के रूप में)	10.6 माइक्रोग्राम/लीटर या कम	
13.	लेड (पीबी के रूप में)	4.6 माइक्रोग्राम/लीटर या कम	
14.	आर्सेनिक (एएस के रूप में)	3.5 माइक्रोग्राम/लीटर या कम	
15.	क्रोमियम (सीआर के रूप में)	8.0 माइक्रोग्राम/लीटर या कम	
16.	मोनाक्रोटोफॉस	89 नैनोग्राम/लीटर या कम	

**मानदंड संबंधी सतत सांद्रण (सीसीसी)–परिवेशी जल में सामग्री के उच्चतम सांद्रण (विषाक्तता की जैव-जांच पर आधारित) का आकलन, जिससे कोई जलीय समुदाय अस्वीकार्य प्रतिकूल प्रभाव डाले बिना अनिश्चित काल के लिए प्रभावित हो सकता है।

(ख) तालिका 1.5—श्रेणी एसडब्ल्यू-V जल क्षेत्र [नेविगेशन और नियंत्रित अपशिष्ट निपटान] के लिए प्राथमिक जल गुणवत्ता मानदंड के नीचे क्रम संख्या 6 से 13 पर क्रम सं. 5 के बाद अतिरिक्त मापदण्ड निम्नानुसार प्रतिस्थापित किए जाएंगे:-

क्र. सं.	मापदंड	मानक	औचित्य/अभ्युक्ति
6.	कैडमियम (सीडी के रूप में)	3.03 माइक्रोग्राम/लीटर या कम	मानदंड संबंधी सतत सांद्रण (सीसीसी) के आधार पर **
7.	तांबा (सीयू के रूप में)	4.1 माइक्रोग्राम/लीटर या कम	
8.	पारा (एचजी के रूप में)	0.38 माइक्रोग्राम/लीटर या कम	
9.	जस्ता (ज़ेडसी के रूप में)	10.6 माइक्रोग्राम/लीटर या कम	
10.	लेड (पीबी के रूप में)	4.6 माइक्रोग्राम/लीटर या कम	
11.	आर्सेनिक (एएस के रूप में)	3.5 माइक्रोग्राम/लीटर या कम	
12.	क्रोमियम (सीआर के रूप में)	8.0 माइक्रोग्राम/लीटर या कम	
13.	मोनाक्रोटोफॉस	89 नैनोग्राम/लीटर या कम	

**मानदंड संबंधी सतत सांद्रण (सीसीसी)–परिवेशी जल में सामग्री के उच्चतम सांद्रण (विषाक्तता की जैव जांच पर आधारित) का आकलन, जिससे कोई जलीय समुदाय अस्वीकार्य प्रतिकूल प्रभाव डाले बिना अनिश्चित काल के लिए प्रभावित हो सकता है।

[फा. सं. क्यू.15017/13/2016-सीपीडब्ल्यू]

जिगमेत टक्पा, संयुक्त सचिव

टिप्पणी: ये मूल नियम भारत के राजपत्र, असाधारण, भाग II, खंड 3, उप-खंड (i) में संख्या का.आ. 844(अ), तारीख 19 नवम्बर, 1986 के द्वारा प्रकाशित किए गए थे और उन्हें अंतिम बार अधिसूचना सा.का.नि. 48(अ), तारीख 24 जनवरी, 2020 के द्वारा संशोधित किया गया था।

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MINISTRY OF ENVIRONMENT, FOREST AND CLIMATE CHANGE

NOTIFICATION

New Delhi, the 17th February, 2020

G.S.R. 122(E).—The following draft of the notification, which the Central Government proposes to issue in exercise of the powers conferred by sections 6 and 25 of the Environment (Protection) Act, 1986 (29 of 1986) is hereby published, as required under sub-rule (3) of rule 5 of the Environment (Protection) Rules, 1986, for the information of the public likely to be affected thereby; and notice is hereby given that the said draft notification shall be taken into consideration on or after the expiry of a period of sixty days from the date on which copies of the Gazette containing this notification are made available to the public.

Any person interested in making any objections or suggestions on the proposals contained in the draft notification may forward the same in writing, for consideration of the Central Government within the period specified above to the Secretary, Ministry of Environment, Forest and Climate Change, Indira Paryavaran Bhawan, Jor Bagh Road, New Delhi-110003, or send it to Member Secretary, CPCB and Scientist 'E' Ministry at the e-mail address i.e. mscb.cpcb@nic.in and h.kharkwal@nic.in.

Draft Notification

The Central Government hereby makes the following rules further to amend the Environment (Protection) Rules, 1986, namely:-

1. **Short title and commencement.**—(1) These rules may be called the Environment (Protection) Amendment Rules, 2020.

(2) They shall come into force on the date of their publication in the Official Gazette.

2. In the Environment (Protection) Rules, 1986, in Schedule-I, for serial number 86 relating to water quality standards for coastal waters marine outfalls, the following entries shall be inserted namely:-

(a) under Table 1.3- Primary Water Quality Criteria for Class SW-III Waters [For Industrial Cooling, Recreation (non-contact) and Aesthetics], serial number 9 to 16, the standards for the additional parameters shall be inserted after Sl.No.8, as follows: -

Sl. No.	Parameter	Standards	Rationale/Remarks
9.	Cadmium (as Cd)	3.03 µg/l or less	Based on Criterion Continuous Concentration (CCC)**
10.	Copper (as Cu)	4.1µg/l or less	
11.	Mercury (as Hg)	0.38µg/l or less	
12.	Zinc (as Zc)	10.6µg/l or less	
13.	Lead (as Pb)	4.6µg/l or less	
14.	Arsenic (as As)	3.5 µg/l or less	
15.	Chromium (as Cr)	8.0µg/l or less	
16.	Monocrotophos	89 ng/l or less	

** Criterion Continuous Concentration (CCC)- an estimate of the highest concentration of the material in ambient water (based on bio-assay toxicity) to which an aquatic community can be exposed indefinitely without resulting in an unacceptable adverse effect.

(b) under Table 1.5-Primary Water Quality Criteria for Class SW-V Waters [For Navigation and Controlled Waste Disposal], serial number 6 to 13, the standards for the additional parameters shall be inserted after Sl.No. 5 as follows: -

Sl. No.	Parameter	Standards	Rationale/Remarks
6.	Cadmium (as Cd)	3.03 µg/l or less	*Based on Criterion Continuous Concentration (CCC)
7.	Copper (as Cu)	4.1µg/l or less	
8.	Mercury (as Hg)	0.38µg/l or less	
9.	Zinc (as Zc)	10.6 µg/l or less	
10.	Lead (as Pb)	4.6µg/l or less	
11.	Arsenic (as As)	3.5µg/l or less	
12.	Chromium (as Cr)	8.0µg/l or less	
13.	Monocrotophos	89 ng/l or less	

* Criterion Continuous Concentration (CCC) - an estimate of the highest concentration of the material in ambient water (based on bio-assay toxicity) to which an aquatic community can be exposed indefinitely without resulting in an unacceptable adverse effect.

[F. No. Q.15017/13/2016-CPW]

JIGMET TAKPA, Jt. Secy.

Note: The principal rules were published in the Gazette of India, Extraordinary, Part II, Section 3, Sub-section (i) *vide* number S.O. 844(E), dated the 19th November, 1986 and lastly amended *vide* notification G.S.R. 48(E), dated the 24th January, 2020.

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64.0 WATER QUALITY STANDARDS FOR COASTAL WATERS MARINE OUTFALLS

In a coastal segment marine water is subjected to several types of uses. Depending of the types of uses and activities, water quality criteria have been specified to determine its suitability for a particular purpose. Among the various types of uses there is one use that demands highest level of water quality/purity and that is termed a "designed best use" in that stretch of the coastal segment. Based on this, primary water quality criteria have been specified for following five designated best uses:-

Class	**Designated best use**
SW-I (see Table 1.1)	Salt pans, Shell fishing, Mariculture and Ecologically Sensitive Zone.
SW-II (see Table 1.2)	Bathing, Contact Water Sports and Commercial fishing.
SW-III(see Table 1.3)	Industrial cooling, Recreation (non-contact) and Aesthetics.
SW-IV (see Table 1.4)	Harbour.
SW-V (see Table 1.5)	Navigation and Controlled Waste Disposal.

The standards along with rationale/remarks for various parameters, for different designated best uses, are given in Table 1.1 to 1.5.

Table 1.1 Primary Water Quality Criteria For Class SW-I Waters
(For Salt pans, Shell fishing, Mariculture and Ecologically Sensitive Zone)

S. No.	Parameter	Standards	Rationale/Remarks
1.	pH range	6.5-8.5	General broad range, conducive for propogation of aquatic lives, is given. Value largely dependant upon soil-water interaction.

(Contd.....)

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2.	Dissolved Oxygen	5.0 mg/l or 60 percent saturation value, whichever is higher.	Not less than 3.5 mg/l at any time of the year for protection of aquatic lives.
3.	Colour and Odour	No noticeable colour or offensive odour.	Specially caused by chemical compounds like creosols, phenols, naptha, pyridine, benzene, toluene etc. causing visible colouration of salt crystal and tainting of fish flesh.
4.	Floating Matters	Nothing obnoxious or detrimental for use purpose.	Surfactants should not exceed an upper limit of 1.0 mg/l and the concentration not to cause any visible foam.
5.	Suspended Solids	None from sewage or industrial waste origin	Settleable innert matters not in such concentration that would impair any usages specially assigned to this class.
6.	Oil and Grease (including Petroleum Products)	0.1 mg/l	Concentration should not exceed 0.1 mg/l as because it has effect on fish egs and larvae.
7.	Heavy Metals: Mercury (as Hg) Lead (as Pb) Cadmium (as Cd)	 0.01 mg/l 0.01 mg/l 0.01 mg/l	Values depend on: (i) Concentration in salt, fish and shell fish. (ii) Average per capita consumption per day. (iii) Minimum ingestion rate that induces symptoms of resulting diseases.

Note : SW-1 is desirable to be safe and relatively free from hazardous chemicals like pesticides, heavy metals and radionuclide concentrations. Their combined (synergestic or antagonistic) effects on health and aquatic lives are not yet clearly known. These chemicals undergo bio-accumulation, magnification and transfer to human and other animals through food chain. In areas where fisheries, salt pans are the governing considerations, and presence of such chemicals apprehended/reported, bioassay test should be performed following appropriate methods for the purpose of setting case-specific limits.

Table 1.2 Primary Water Quality Criteria for Class SW-II Waters

(For Bathing, Contact Water Sports and Commercial Fishing)

S. No.	Parameter	Standards	Rationale/Remarks
1.	pH range	6.5-8.5	Range does not cause skin or eye irritation and is also conducive for propagation of aquatic life.
2.	Dissolved Oxygen	4.0 mg/l or 50 percent saturation value whichever is higher.	Not less than 3.5 mg/l at anytime for protection of aquatic lives.
3.	Colour and Odour	No noticeable colour or offensive odour.	Specially caused by chemical compounds like creosols phenols, naptha, benzene pyridine, volume etc. causing visible colouration of water and tainting of and odour in fish flesh.
4.	Floating Matters	Nothing obnoxious or detrimental for use purpose.	None in concentration that would impair usages specially assigned to this class.
5.	Turbidity	30 NTU (Nephelo Turbidity Unit)	Measured at 0.9 depth.
6.	Fecal Coliform	100/100 ml (MPN)	The average value not exceeding 200/100 ml. in 20 percent of samples in the year and in 3 consecutive samples in monsoon months.
7.	Biochemical Oxygen Demand (BOD) (3 days at 27°C)	3 mg/l	Restricted for bathing (aesthetic quality of water). Also prescribed by IS:2296-1974.

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Table 1.3 Primary Water Quality Criteria for Class SW-III Waters
[For Industrial cooling, Recreation (non-contact) and Aesthetics]

S. No.	Parameter	Standards	Rationale/Remarks
1.	pH range	6.5-8.5	The range is conducive for propagation of aquatic species and restoring natural system.
2.	Dissolved Oxygen	3.0 mg/l or 40 percent saturation value which-ever is higher.	To protect aquatic lives.
3.	Colour and Odour	No noticeable colour or offensive odour.	None in such concentration that would impair usages specifically assigned to this class.
4.	Floating Matters	No visible/obnoxious floating debris, oil slick, scum.	As in (3) above.
5.	Fecal Coliform	500/100 ml (MPN)	Not exceeding 1000/100 ml in 20 percent of samples in the year and in 3 consecutive samples in monsoon months.
6.	Turbidity	30 NTU	Reasonably clear water for Recreation, Aesthetic appreciation and Industrial cooling purposes.
*7.	Dissolved Iron (as Fe)	0.5 mg/l or less	It is desirable to have the collective concentration of dissolved Fe and Mn less or equal to 0.5 mg/l to avoid scaling effect.
*8.	Dissolved Manganese (as Mn)	0.5 mg/l or less	

* Standard included exclusively for Industrial Cooling purpose. Other parameters same.

Table 1.4 Primary Water Quality Criteria for Class SW-IV Waters
(For Harbour Waters)

S. No.	Parameter	Standards	Rationale/Remarks
1.	pH range	6.5-9.0	To minimize corrosive and scaling effect.
2.	Dissolved Oxygen	3.0 mg/l or 40 percent saturation value whichever is higher	Considering bio-degradation of oil and inhibition to oxygen production through photosynthesis.
3.	Colour and Odour	No visible-colour or offensive odour.	None from reactive chemicals which may corrode paints/metallic surfaces.
4.	Floating materials Oil, grease and scum (including Petroleum products)	10 mg/l	Floating matter should be free from excessive living organisms, which may clog or coat operative parts of marine vessels/equipment.
5.	Fecal Coliform	500/100 ml (PAN)	Not exceeding 1000/100 ml in 20 percent of samples in the year and in 3 consecutive samples in monsoon months.
6.	Biochemical Oxygen Demand (3 days at 27°C)	5 mg/l	To maintain water relatively free from pollution caused by sewage and other decomposable wastes.

Table 1.5 Primary Water Quality Criteria for Class SW-V Waters
(For Navigation and Controlled Waste Disposal)

S. No.	Parameter	Standards	Rationale/Remarks
1.	pH range	6.0-9.0	As specified by New England Interstate Water Pollution Control Commission.
2.	Dissolved Oxygen	3.0 mg/l or 40 percent saturation value which ever is higher	To protect aquatic lives.

(Contd.....)

(Contd.....)

3.	Colour and Odour	None is such concentration that would impair any usages specifically assigned to this class.	As in (1) above
4.	Sludge deposits, Solid refuse floating oil, grease & scum.	None except for such small solids, amount that may result from discharge of appropriately treated sewage and/or individual waste effluents.	As in(1) above
5.	Fecal Coliform	500/100 ml (MPN)	Non exceeding 1000/100 ml in 20 percent of samples in the year and in 3 consecutive samples in monsoon months.

Source : EPA, 1986
[GSR 7, dated Dec. 22, 1998]

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ORIGINAL ARTICLE

Bioaccumulation of heavy metals in seafood resources from the southwest coast of India: human health risk assessment and importance of seafood security

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Abstract

Objectives The present study was to evaluate the bioaccumulation of heavy metals and possible human health risk from contaminated seafood resources from the Cochin Estuarine System, Southwest coast of India.

Methods Water, sediment, fish, and shellfish samples were collected from the Cochin Estuarine System during the monsoon and non-monsoon period and were analyzed for heavy metals. Human health risk from contaminated fishes was analyzed using deterministic and probabilistic risk assessment methods.

Results Water, sediment, and biota exhibited elevated concentrations of heavy metals during non-monsoon periods. Zinc was the most abundant metal, while cadmium had the lowest concentration among the three mediums. The presence of cadmium in the estuary posed a moderate to very high ecological risk. Zinc, cadmium, and chromium exceeded the toxicity threshold outlined in the sediment quality guidelines. All metals except cadmium showed significant variation between species and season ($p < 0.05$). Chromium and zinc correlated positively with fish size, while Cu, Cd, Mn, and Ni showed a negative correlation ($p < 0.05$). Chromium had the highest accumulation in herbivores, and Cd was highest in filter feeders. Cadmium and lead exceeded the standards of the Food Safety and Standards Authority of India and the Food and Agriculture Organization. The estimated daily intake and target hazard quotient of individual metals were found to be very low. In the Monte Carlo simulation, the 95th percentile of the hazard index (0.80) indicates that consumers are close to experiencing non-carcinogenic health risks. The cancer risk for cadmium (2.15E−3) and chromium (3.90E−4) suggests that consumers may be vulnerable to long-term risk. Cadmium was found to be the most influencing metal in the risk analysis.

Conclusion The results indicate the need for regular monitoring and risk assessment of heavy metals, especially lead, cadmium, and chromium, in seafood resources across coastal areas to ensure food security.

Keywords Urban estuary · Heavy metals · Fishery resource · Human health risk · Monte Carlo simulation

Introduction

The global challenge of heavy metal pollution in aquatic ecosystems is growing more severe. These metals pose a threat to aquatic life by persisting in their habitats and building up in their bodies, leading to physiological and biochemical damage that could ultimately cause mortality or long-term harm to future generations [1, 2]. Moreover, the detrimental impact of these elements extends to humans who consume contaminated food, which leads to harmful consequences on human health, such as kidney failure, liver damage, cardiovascular diseases, breast cancer, and other carcinogenic effects [3]. Fish and shellfish play a crucial

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role in transferring heavy metals to humans [4]. Despite the potential risks associated with contaminants, fish consumption is increasing globally as people become more aware of its nutritional benefits. As rapid economic and social advancements unfold globally and regionally, heavy metals accumulate within various ecosystems and food chains. This leaves a lasting impact, particularly in developing countries like India, where a substantial portion of coastal communities rely on seafood resources. Therefore, comprehending the levels of heavy metals within the food chain allows us to assess their transmission to humans through the consumption of seafood [4]. Gathering data on the types of fish consumed, as well as the extent of heavy metal contamination, can prove advantageous in reducing public health risks [5]. Several studies indicate that the concentrations of heavy metals in seafood intended for human consumption exceed the thresholds established by international organizations, underscoring potential health hazards for consumers [6–9].

Southern India harbors numerous estuarine and marine ecosystems, among which the Cochin estuarine system (CES), situated in the state of Kerala, stands out as one of the largest estuarine system, which sustains exceptional biodiversity and functions as a crucial breeding and nursery habitat for diverse fish and shellfish species [10]. It constitutes the northern segment of the Vembanad estuary, designated as a Ramsar site in India, with an annual landing of 3906.33 tons [11]. Fishing has been a vital source of livelihood for the residents along the estuary. The fishery resources originating from CES are highly sought after in domestic and international markets, contributing significantly to the economy [12]. However, the CES faces increasing pressure from commercial and industrial activities, with approximately 95% of red-category industries concentrated in Greater Kochi, located within the northern limb of the estuary [10]. Furthermore, contaminants from adjoining Periyar river exacerbate coastal water pollution, posing threat to fishery resources and other marine life. According to [13], the lower Periyar has been described as a cesspool of toxins with alarming levels of heavy metal pollution. Various studies have underscored the severity of heavy metal pollution in the water and sediment of this area [14–16]. However, there is a research gap in regarding the bioaccumulation of heavy metals and the associated human health risks. In order to mitigate potential negative impacts on human health, it is essential to continuously monitor and evaluate the levels of heavy metals in fish and shellfish, which serve as significant components of the local food chain.

In this context, this pioneering report from Kerala includes a comprehensive examination of heavy metal bioaccumulation in commonly consuming seafood resources and associated carcinogenic and non-carcinogenic risk assessment for public health. These findings are crucial for crafting monitoring strategies, enacting corrective actions, and fostering future research pursuits. It provides fundamental data for coastal nations to undertake additional relevant studies and formulate policy-making initiatives directed toward ensuring food security and conserving the coastal ecosystem.

Results

Heavy metals in water and sediment matrix

The dissolved metal concentration in CES followed the order Zn (60.95 ± 69.58 μg L^{-1}) > Pb (5.46 ± 3.6 μg L^{-1}) > Ni (3.07 ± 2.15 μg L^{-1}) > Mn (3.01 ± 2.95 μg L^{-1}) > Cu (2.39 ± 1.29 μg L^{-1}) > Cr (1.5 ± 1.25 μg L^{-1}) > Cd (1.23 ± 1.16 μg L^{-1}). The metals Cu, Zn, Pb, Cd, Ni, and Cr in water showed the highest concentration at the northern estuary (Fig. 1a and Table 1). Seasonally, all the metals showed higher concentrations during the premonsoon (PRM) and postmonsoon (POM) periods than in the monsoon (MON) period (Fig. 1c). The concentration of dissolved Cd showed significant seasonal variation ($p < 0.05$).

The sediment bound metal in CES followed the order Zn (106 ± 99.63 mg kg^{-1}) > Mn (102.39 ± 65.19 mg kg^{-1}) > Cr (64.54 ± 33.92 mg kg^{-1}) > Ni (19.44 ± 11.52 mg kg^{-1}) > Cu (15.37 ± 8.13 mg kg^{-1}) > Pb (8.53 ± 5.13 mg kg^{-1}) > Cd (2.69 ± 2.52 mg kg^{-1}). The sediment exhibited the highest concentrations of metals (Cu, Zn, Mn, Ni, and Cr) within the northern estuary (Fig. 1b and Table 1). It displayed higher concentrations during the non-monsoon season compared to the monsoon season (Fig. 1d). Cadmium and Cr displayed significant seasonal variations, whereas Zn exhibited significant spatial variation ($p < 0.05$). The shade plot shows the distribution and concentration of metals in sediment and water within the CES, highlighting their contribution to variations among distinct group (Figs. S1 and S2). Elevated concentrations of Zn, Mn, and Cr were observed in sediment, making them prominent metals influencing differences among stations. Conversely, water analysis reveals higher levels of Zn, Pb, Ni, and Mn, identifying them as major metals influencing the variations among stations.

Various sediment quality indices were utilized to assess the extent of heavy metal pollution in the estuarine sediment. The geo-accumulation index indicates that Cd contamination was moderate in the central and northern estuary. Zinc levels ranged from uncontaminated to moderately contaminated in both the central and northern estuary (Fig. S3a and Table S1). The contamination factor indicates moderate Zn contamination in the central and northern estuary, while a considerable contamination factor was observed for Cd in both the central and northern estuary (Fig. S3b and Table S1). The ecological risk level associated with each individual heavy metal can be assessed based on its

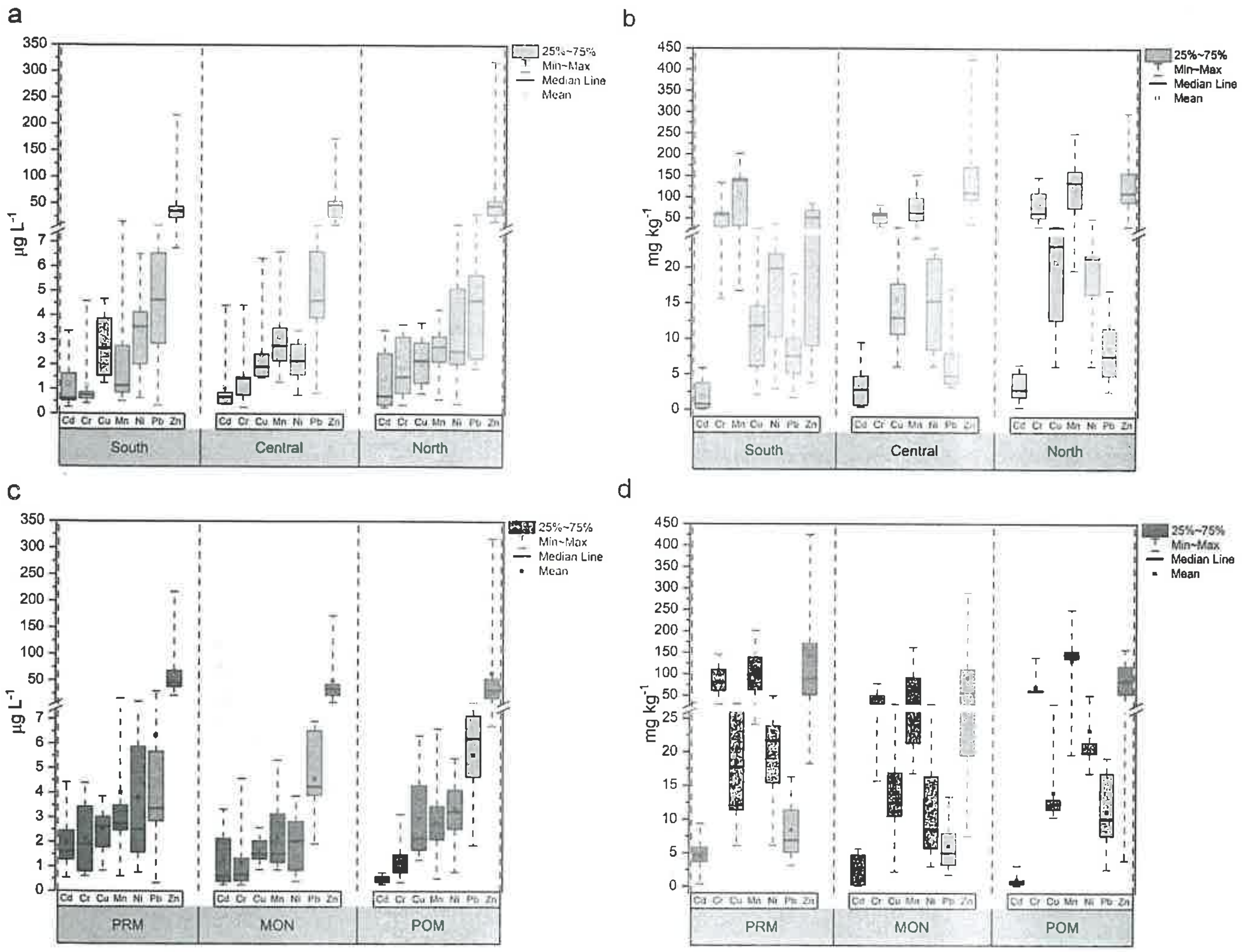

Fig. 1 Heavy metal distribution in water and sediment of CES **a** Zone-wise metal concentration in water **b** Zone-wise metal concentration in sediment **c** Seasonal metal concentration in water **d** Seasonal metal concentration in sediment

Table 1 Heavy metal concentration in water and sediment samples of Cochin estuarine system during the study period

	Cu	Zn	Pb	Cd	Mn	Ni	Cr
Water ($\mu g\ L^{-1}$)							
Min	0.83	6.75	0.35	0.25	15.85	0.41	0.25
Max	6.34	318.56	30.15	4.41	8.85	10.15	4.58
Mean ± SD	2.39 ± 1.29	60.95 ± 69.58	5.46 ± 3.60	1.23 ± 1.16	3.01 ± 2.95	3.07 ± 2.15	1.5 ± 1.25
Sediment ($mg\ kg^{-1}$)							
Min	2.12	3.78	1.65	0.018	16.73	2.95	15.61
Max	31.1045	424.85	19.047	9.374	248.92	48.91	145.67
Mean ± SD	15.37 ± 8.13	106 ± 99.63	8.53 ± 5.13	2.69 ± 2.52	102.39 ± 65.19	19.44 ± 11.52	64.54 ± 33.92

respective potential risk factor. In the present investigation, Cd demonstrated a range from moderate to very high ecological risk. In contrast, all other metals indicated a low ecological risk within the estuary (Fig. S3c and Table S2). The Potential Ecological Risk Index (PERI) values suggest that the central and northern estuary was experiencing a moderate ecological risk (Fig. S3d and Table S2). In the southern, central, and northern estuary, Cu exceeded the threshold effect level (TEL), maximum concentration at which no toxic effects are observed. Zinc surpassed the probable effect level (PEL) in the northern estuary and the effects range median (ERM) in the central estuary. Cadmium level exceeded the PEL in the southern and northern estuary and the ERM in the central estuary. Nickel concentrations surpassed the effects range low (ERL) in the southern, central, and the northern estuary. Chromium exceeded the ERL in the southern, central, and northern estuary. Notably, Zn, Cd, and Cr levels in the estuary surpassed the higher toxicity thresholds (PEL and ERM) (Table S3).

Heavy metal bioaccumulation in fishes

The annual average bioaccumulation of Cu during the study period was 7.05 ± 6.44 mg kg^{-1} and the highest Cu accumulation was in *Caranx ignobilis* and lowest was in *Portunus pelagicus* (Fig. 2a, b). Seasonally, the highest accumulation of Cu was observed during POM (10.68 ± 7.10 mg kg^{-1}), with the lowest in *P. pelagicus* and the highest in *C. ignobilis* (Fig. 2c and Fig. S4 a–c). Zonally, highest was in the northern zone, measured at 7.51 ± 6.79 mg kg^{-1}, and the lowest in the central zone, recorded as 6.50 ± 6.51 mg kg^{-1} (Fig. 2d).

The annual mean concentration of Zn was 13.28 ± 13.70 mg kg^{-1}, with lowest in *Mugil cephalus* and highest in *Villorita cyprinoides* (Fig. 2a & b). Seasonally, the highest accumulation was noted during PRM (16.55 ± 17.04 mg kg^{-1}), with the lowest in *Penaeus monodon* and the highest in *V. cyprinoides* (Fig. 2c and Fig.S4 a–c). Zonally, it was highest in the central (14.73 ± 15.44 mg kg^{-1}) and lowest in the northern zone (11.95 ± 13.10 mg kg^{-1}) (Fig. 2d). Overall, Zn concentration was the highest among all metals across all species.

On an annual basis, Pb demonstrated an accumulation of 0.53 ± 0.61 mg kg^{-1} (Fig. 2a). *C. ignobilis* exhibited the lowest accumulation, while the highest accumulation was observed in *M. cephalus* (Fig. 2b). Season-wise, the highest accumulation was observed during POM (0.62 ± 0.68 mg kg^{-1}), with the lowest in *V. cyprinoides*

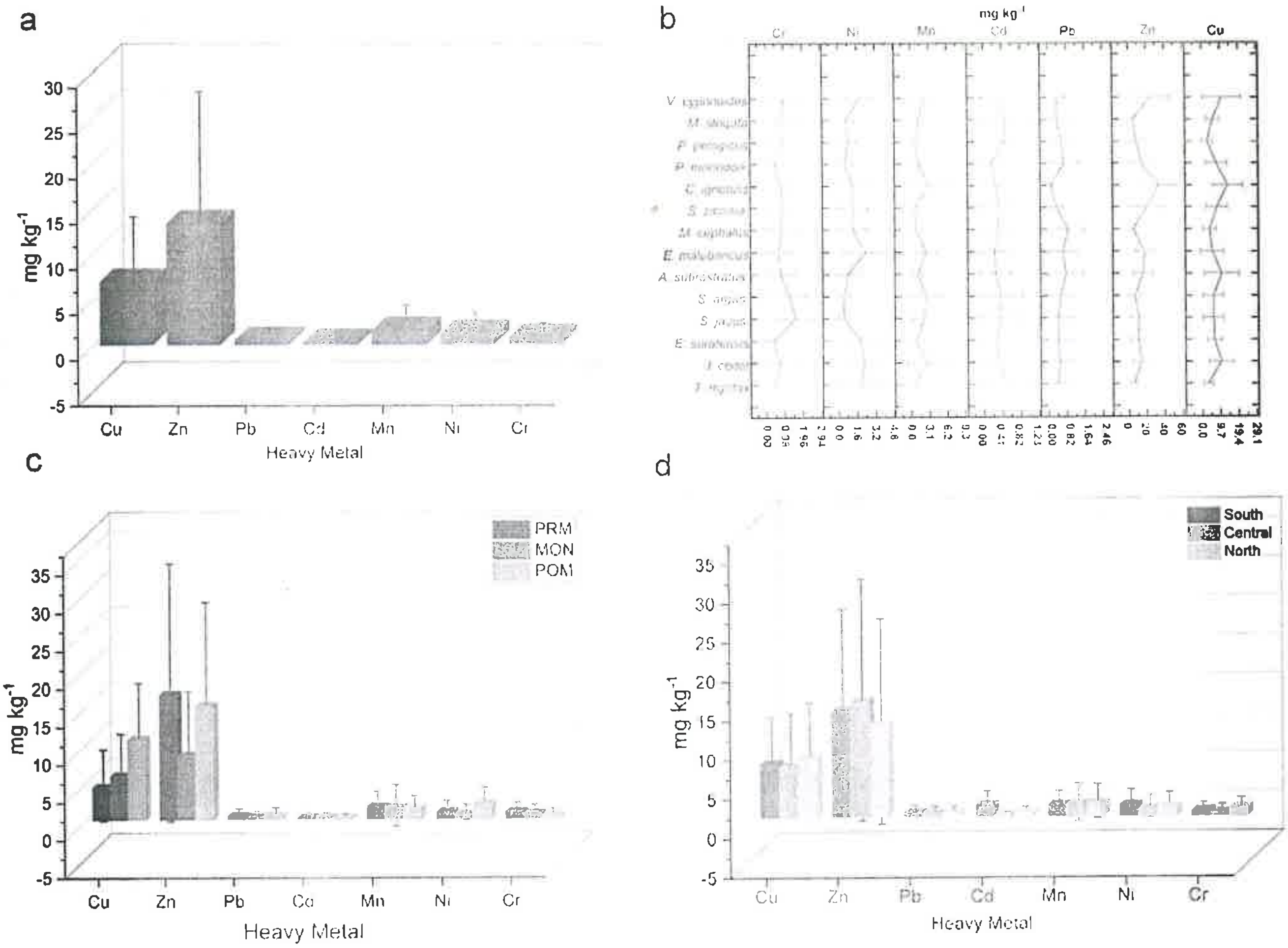

Fig. 2 Heavy metal concentration in fish and shellfish species from CES **a** Annual metal accumulation in CES **b** Mean metal concentration in individual species (mg kg dw^{-1}) **c** Seasonal metal accumulation in CES **d** Zone-wise metal accumulation in CES

and the highest in *Arius subrostratus* (Fig. 2c and Fig. S4 a-c). Zone-wise, it showed elevated concentration in the southern zone (0.54 ± 0.62 mg kg^{-1}) and lower concentration in the central zone (0.48 ± 0.60 mg kg^{-1}) (Fig. 2d).

Cadmium exhibited an annual accumulation of 0.31 ± 0.39 mg kg^{-1}, with highest in *P. monodon* and the lowest in *Thryssa mystax* (Fig. 2a & b). Seasonally, Cd displayed its peak accumulation during PRM (0.33 ± 0.48 mg kg^{-1}), with the lowest concentration observed in *C. ignobilis* and the highest in *Johnius coitor* (Fig. 2c and Fig. S4a–c). Zone-wise it showed highest at northern zone (0.42 ± 0.56 mg kg^{-1}) and lowest in the central zone (0.24 ± 0.19 mg kg^{-1}) (Fig. 2d).

Manganese displayed an average annual accumulation of 1.58 ± 2.03 mg kg^{-1}, and *T. mystax* exhibited the lowest accumulation, while *C. ignobilis* showed the highest (Fig. 2a, b). Seasonally, Mn exhibited its highest accumulation during PRM (1.73 ± 1.74 mg kg^{-1}), with the lowest observed in *Sillago sihama* and the highest in *Sactophagus argus* (Fig. 2c and Fig. S4 a–c). Zonally, highest concentration was found in the northern zone (1.68 ± 2.14 mg kg^{-1}), while the southern zone showed the lowest (1.52 ± 1.50 mg kg^{-1}) (Fig. 2d).

Nickel showed an annual average accumulation of 1.27 ± 1.54 mg kg^{-1} (Fig. 2a). The lowest concentration was found in *S. javus* and highest in *E. malabaricus* (Fig. 3b). Seasonally, it showed highest accumulation in POM (1.96 ± 1.95 mg kg^{-1}) and lowest in *Mytella strigata* (0.59 ± 0.70 mg kg^{-1}) and highest in *T. mystax* (4.56 ± 1.69 mg kg^{-1}) (Fig. 2c and Fig. S4 a–c). Zonally, highest concentration was in the southern estuary (1.48 ± 1.66 mg kg^{-1}) and lowest in the central estuary (0.99 ± 1.44 mg kg^{-1}) (Fig. 2d).

Chromium exhibited an annual average accumulation concentration of 0.80 ± 0.85 mg kg^{-1}, and the lowest accumulation was observed for *E. suratensis*, while the highest was found in *S. javus* (Fig. 2a, b). Seasonally, Cr displayed its highest concentration during PRM (1.04 ± 0.94 mg kg^{-1}), with lowest accumulation in *C. ignobilis* and highest in *S. argus* (Fig. 2c and Fig. S4 a–c). Zonally, northern estuary showed highest accumulation (1.11 ± 0.97 mg kg^{-1}), while the central estuary shows the lowest (0.54 ± 0.69 mg kg^{-1}) (Fig. 2d).

The order of the mean metal accumulation in CES during the study period was Zn > Cu > Mn > Ni > Cr > Pb > Cd. All the metals except Cd showed statistically significant variation between species and season ($p < 0.05$). Copper, Zn, Pb, Cd, and Mn showed statistically significant variation between zones ($p < 0.05$). The seasonal and zone-wise metal concentration in species is illustrated in Fig. S4a–f.

The Pearson correlation test revealed significant relationships between metal concentrations and biological parameters across various fish species. Copper showed a negative correlation with length ($r = -0.382$, $p = 0.049$) and weight ($r = -0.687$, $p = 0.00$) of *C. ignobilis*. Zinc showed a positive relationship with length ($r = 0.482$, $p = 0.011$) and weight ($r = 0.468$, $p = 0.014$) of *C. ignobilis*. Negative size-dependent relationships were found for Mn in *Etroplus suratensis* ($r = -0.388$, $p = 0.046$) and *Epinephelus malabaricus* ($r = -0.397$, $p = 0.049$). Cadmium negatively correlated with length in *P. pelagicus* ($r = -0.388$, $p = 0.045$). Nickel showed a negative relationship in *P. pelagicus* ($r = -0.440$, $p = 0.022$) and *V. cyprinoides* ($r = 0.679$, $p = 0.044$). Chromium showed significant positive correlation with length ($r = 0.485$, $p = 0.010$) and weight ($r = 0.561$, $p = 0.002$) of *P. monodon* (Fig. 3 and Tables S4 and S5).

Human health risk assessment

Deterministic risk assessment

Estimated daily intake (EDI)

The estimated daily intake (EDI) serves as a vital metric for gauging the average daily influx of metals into the human body, providing essential insights into potential exposure levels and their implications on human health. In the present study, the highest EDI was observed for Zn in *C. ignobilis*, while the lowest was for Pb in the same species. All metal EDIs remained under the suggested daily intake (RfD), and it showed the following decreasing order Zn > Cu > Mn > Ni > Cr > Pb > Cd (Table 2).

Target hazard quotient (THQ) and hazard index (HI)

The assessment of potential health risks associated with exposure to various contaminants heavily relies on the target hazard quotient (THQ) and hazard index (HI), offering essential insights into the management of environmental and human health concerns. In this investigation, Cd demonstrated the highest THQ (0.25 ± 0.08), ranging from 0.12 (*P. monodon*) to 0.37 (*T. mystax*), while Mn exhibited the lowest THQ ($8.82 \times 10^{-3} \pm 6.76 \times 10^{-3}$), with a range of 2.9×10^{-3} (*T. mystax*) to 2×10^{-1} (*C. ignobilis*). The THQ values for the metals followed the sequence: Cd > Cr > Cu > Pb > Ni > Zn > Mn (Fig. 4a, b). In terms of the HI, the highest value was observed for *S. javus* (0.95), whereas the lowest was for *P. monodon* (0.60), with an average of 0.67 ± 0.13 (Fig. 4c).

Target cancer risk (TCR)

The target cancer risk (TCR) for Pb ranged from 8.15×10^{-7} in *C. ignobilis* to 5.7×10^{-6} in *M. cephalus*. For Cd, it varied between 7.54×10^{-4} in *S. argus* and 2.35×10^{-3} in *M.*

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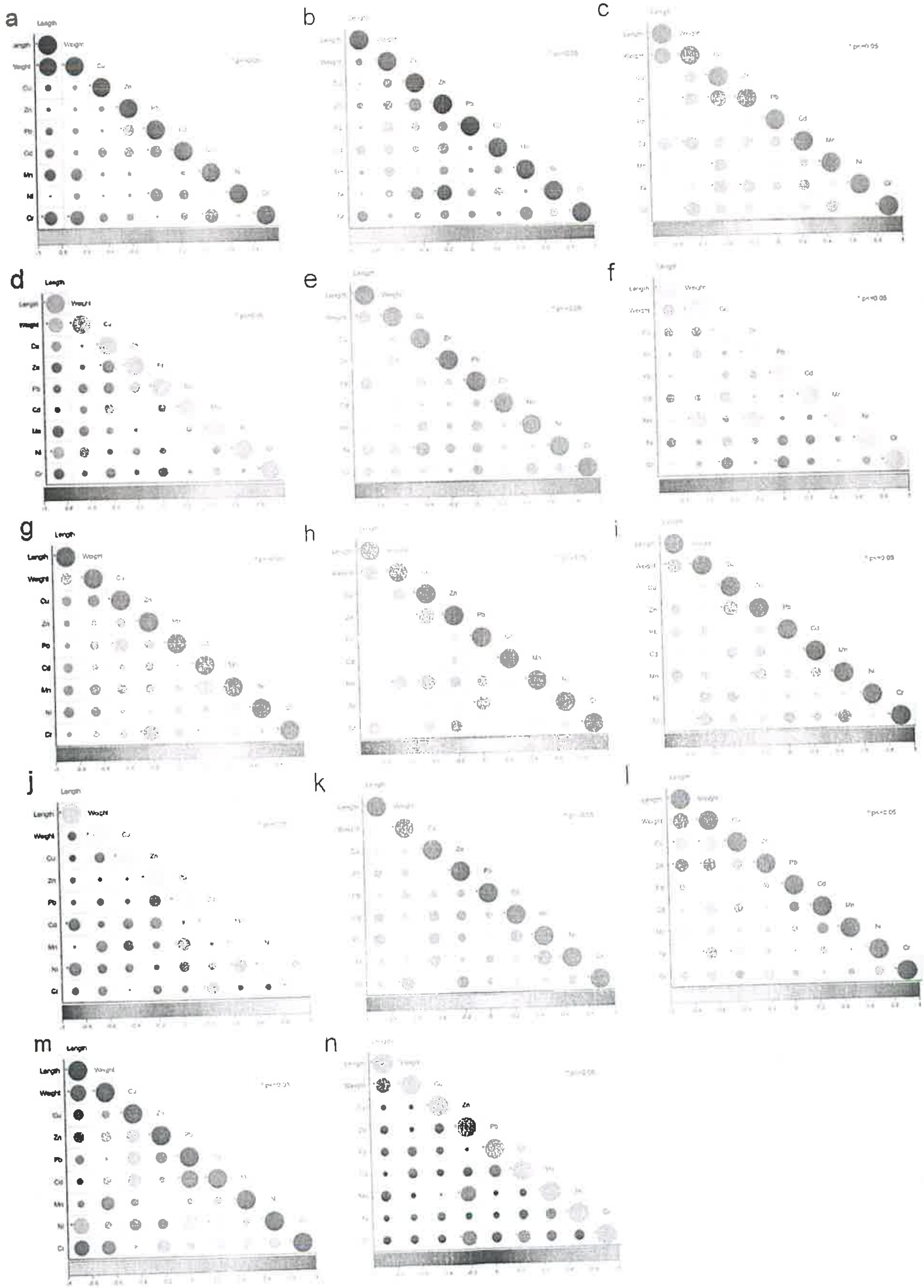

◂**Fig. 3** Correlation plot of metal concentration with length and weight **a** *P. Monodon* **b** *T. Mystax* **c** *J. Coitor* **d** *E. Suratensis* **e** *M. Strigata* **f** *S. Argus* **g** *A. subrostratus* **h** *E. Malabaricus* **i** *M. cephalus* **j** *P. pelagicus* **k** *S. Sihama* **l** *C. ignobilis* **m** *V. cyprinoides* **n** *S. javus*

strigata. Regarding Cr, it spanned from 1.38×10^{-4} in *E. suratensis* to 5.98×10^{-4} in *S. javus*. Cadmium presented a carcinogenic risk in 87% of the organisms studied, with the remainder falling within acceptable limits (10^{-4}). The TCR for Cr suggested acceptable levels in the majority of organisms, with negligible health risk only evident in *J. coitor* and *E. suratensis*. For Pb, the TCR remained within an acceptable limit in most species and exhibited negligible risk in *C. ignobilis*. The observed order of TCR was Cd > Cr > Pb (Table 3).

Probabilistic risk assessment and sensitivity analysis

The mean and 95th percentile values of HI by probabilistic approach were 0.38 and 0.80, respectively (Fig. 5a). For TCR of Pb, they were 1.95×10^{-6} and 6.54×10^{-6} respectively. The TCR of Cd showed a mean value of 7.06×10^{-4}, and the 95th percentile value was 2.15×10^{-3}. The mean simulated TCR for Cr was, 1.15×10^{-4}, and at the 95th percentile, it was 3.90×10^{-4} (Fig. 5b–d). The sensitivity analysis demonstrated that the non-carcinogenic risk is primarily affected by metal concentration (70.22%), with exposure duration also playing a significant role (20.09%). Among the metals analyzed, Cd exhibited the highest sensitivity, accounting for 24.58%, followed by Pb at 18.62%. In terms of cancer risk, Pb, Cd, and Cr individually contributed 93.90%, 91.93%, and 94.53%, respectively (Fig. 6a–d).

Discussion

Fish and shellfish, spanning various trophic levels, often serve as key indicators of aquatic pollution [17]. Pollution in aquatic habitats introduces contaminants into human diets through the food chain, stressing the need to quantify the contaminants in them since their consumption is very important for the nutrition and growth of human beings. This study, therefore, focused on heavy metal bioaccumulation in finfish and shellfish from an urban estuary on the southwest coast of India, coupled with human health risk assessment. There are many studies on the distribution of heavy metals in water and sediment from CES, but little is known about bioaccumulation trends in the estuary. Additionally, despite the significance of fish in the Keralite diet, comprehensive investigations into human health risks stemming from heavy metal-contaminated diets still need to be explored in Kerala.

All samples analyzed in the study were sourced from the commercial fishery catch, representing native, edible species with high consumer demand in Kerala. Heavy metal distribution across three sampling zones revealed that fish from the northern zone had the highest metal accumulation compared to the central and southern zones. This aligns with previous findings of elevated metal concentrations in sediment and water on the northern side of the estuary [14, 15, 18]. Non-monsoonal periods exhibited higher tissue accumulation rates compared to the monsoon season. This heightened accumulation during dry seasons can be attributed to increased metabolic activity and feeding cycles of fish [19]. In addition to this factor, another cause may be that heavy metals increase their concentration in dry seasons, so that organisms tend to show higher concentrations during this season. Variability in heavy metal bioaccumulation across seasons can also be explained by shifts in estuarine pollution levels. During the monsoon season, substantial rainfall and heightened river flow led to reduction in metal concentration within the CES [14]. In the present study, also the metal concentration in CES water was highest in PRM, followed by POM and MON. Dissolved metal concentration was higher toward the northern parts of the estuarine system, where most of the industries are located and this is in agreement with the previous studies by [15, 16]. Sediment metal concentration also showed higher level during the dry period. The presence of high heavy metal content in muscle tissue observed in the study during non-monsoonal periods aligns with these findings.

The heavy metal level in water and sediment fractions of CES were compared with that of other regions of the world (Table S6) and all the dissolved metals in CES showed higher concentrations compared to other estuaries. Sediment-bound Cd and Cr showed higher values than the world average shale in Earth's crust. Copper, Cd, Ni, and Cr exceeded the toxicity reference value, which indicates that a dietary contaminant dose may pose a potential risk to a predatory ecological receptor. In sediments of CES, toxic metal Pb was found to be higher than that in the Uppanar estuary (India), the Yellow River Estuary (China), and the Arabian Gulf coast. Similarly, Cd contamination was found to be much higher than that in the Biyyam Backwater (Kerala), the Uppanar Estuary, the Malthuze Estuary (South Africa), the Yellow River Estuary (China), the Daya Bay (China), and the Yellow Sea (South Korea) (Table S6). During the study period, the levels of Zn, Cd, and Cr in the sediment exceeded higher toxicity thresholds, suggesting that contamination from these metals can have notable adverse effects.

Many studies have reported a positive correlation between metal accumulation and fish size; this investigation uncovered both positive and negative correlations. Varying stress responses, physiological conditions, and detoxification mechanisms of the individual fish may also influence the

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Table 2 Estimated daily intake of heavy metals (μg kg bw^{-1} day^{-1}) from the fish and shellfish from CES and its comparison with RfD

Organism	Cu	Zn	Pb	Cd	Mn	Ni	Cr
T. mystax	2.39	4.30	0.32	0.37	0.41	1.45	0.33
J. coitor	7.62	10.70	0.42	0.26	2.05	1.70	0.54
E. suratensis	4.96	7.86	0.36	0.22	0.63	1.44	0.28
S. javus	4.48	8.47	0.34	0.29	1.70	0.38	1.20
S. argus	4.47	5.21	0.42	0.28	1.87	0.38	0.96
A. subrostratus	7.91	12.05	0.60	0.20	0.84	0.62	0.60
E. malabaricus	4.15	13.25	0.50	0.18	1.97	1.80	0.51
M. cephalus	2.97	3.47	0.67	0.24	0.62	0.96	0.61
S. sihama	5.96	15.76	0.27	0.25	0.48	0.97	0.60
C. ignobilis	10.44	25.81	0.10	0.20	2.47	0.94	0.62
P. monodon	5.84	11.29	0.53	0.17	0.93	0.44	0.36
P. pelagicus	2.08	6.82	0.48	0.35	0.56	0.69	0.68
M. strigata	4.37	3.86	0.29	0.37	0.97	0.52	0.59
V. cyprinoides	8.06	18.83	0.31	0.20	1.76	1.42	0.74
RfD	40	300	3.5	1	140	20	3

body metal burden other than the length and weight [20, 21]. Hence, this study recommends a comprehensive analysis of the relationship between allometry and metal accumulation, considering the influence of stress responses in organisms from the field as well as through laboratory experiments.

Bioaccumulation data cited in Table S7 show that more than 50% of the literature reported lower metal concentrations for all the metals compared to this investigation. Copper concentration in this study was comparable to those in the Ennore estuary [22] but exceeded values from other regions of Kerala [23, 24]. Mean Zn concentration in our study was lower than those reported from Kerala [23, 24] and higher than in many estuaries from India and Bangladesh [25–28]. Lead and Cd concentrations were lower than in some studies [24, 28] but higher than in [26], while Mn showed a higher distribution compared to [3, 29] and lower compared to [23, 26]. Even though Mn causes serious toxicity effects, studies on metal bioaccumulation are very much limited. Nickel concentration was higher than in [26, 30], and Cr was within a comparable range reported from Visakhapatnam [31]. As these heavy metals cause cardiovascular toxicity, hepatotoxicity, infant mortality, and neurological complications like Alzheimer's and Parkinson's disease, lung cancer, and oxidative stress [29], regular monitoring and risk assessment should be carried out to enforce strict laws, regulations, and water quality guidelines in the future. Also, the metals Pb, Cd, and Cr have exceeded the standard limits set by international and national organizations like the Food Safety and Standards Authority of India, the Joint FAO/WHO Expert Committee on Food Additives, the Food and Drug Administration, and the Federal Environmental Protection Agency (Fig. S5 and Table S8).

The metals Cu, Zn, Pb, Mn, and Ni showed highest accumulation in carnivorous species, while Cr was highest in herbivores and lowest in carnivores. Cadmium showed the highest levels in filter feeders and the lowest in detritivores. Hence, this study indicates there is no discernible trophic relationship in the bioaccumulation pattern within the CES. This suggests that factors beyond feeding habits and species-specific variations in bioaccumulation may be influenced by physiological traits, specific mechanisms for metal accumulation, variations in metabolism and absorption, the metal content in the feed, and the rate of biomagnification [32].

To assess human health risks associated with studied heavy metals, EDI, THQ, and HI were computed. Estimated daily consumption is used to calculate the dietary exposure to heavy metals for an individual who consumes fish in their regular diet. The EDI values were well below the recommended daily allowance standards, demonstrating the low risk of these metals pose to consumer health. The potential health risks linked to long-term dietary metal consumption were estimated using the target hazard quotient. Hazard index is the sum of THQ values for each metal. The THQ values of all the metals were well below the threshold (1), suggesting that health effects associated with Cu, Zn, Pb, Cd, Mn, Ni, and Cr are unlikely to occur. But, the HI values are very near the threshold value, which shows that humans are not far away from the synergistic effects of these metals. Previously non-carcinogenic risk of Cu and Zn was assessed by [33] on the fish *E. suratensis* collected from the CES using THQ only, which showed no health risk on consuming this fish from CES. Compared to [33], the present investigation used HI in conjunction with THQ for a better understanding of risk from synergistic effects of metals.

Probabilistic risk assessment is crucial for enhancing the precision of health risk assessment results [34]. The average levels of HI are below one and do not pose any health hazards to the public. In Monte Carlo simulation-based

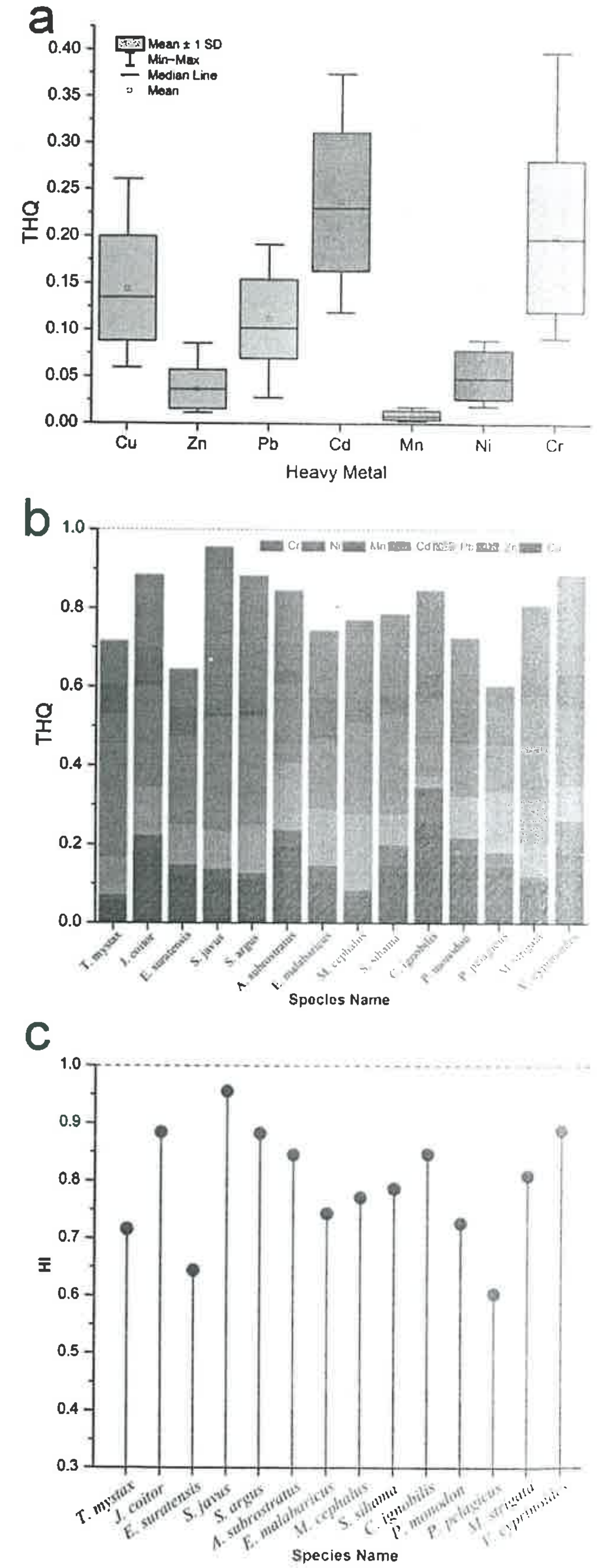

Fig. 4 Non-carcinogenic risk of heavy metals for consumers. **a** Mean target hazard quotient **b** THQ in individual species **c** Hazard index

Table 3 TCR of Pb, Cd, and Cr for different species

Species	Pb	Cd	Cr
T. mystax	2.76E–06	2.30E–03	1.63E–04
J. coitor	3.54E–06	1.63E–03	2.72E–04
E. suratensis	3.02E–06	1.41E–03	1.38E–04
S. javus	2.86E–06	1.82E–03	5.98E–04
S. argus	3.6E–06	1.76E–03	4.81E–04
A. subrostratus	5.08E–06	1.26E–03	2.99E–04
E. malabaricus	4.26E–06	1.12E–03	2.55E–04
M. cephalus	5.7E–06	1.49E–03	3.06E–04
S. sihama	2.33E–06	1.60E–03	3.01E–04
C. ignobilis	8.16E–07	1.27E–03	3.10E–04
P. monodon	4.54E–06	7.53E–04	1.79E–04
P. pelagicus	4.12E–06	2.24E–03	3.40E–04
M. strigata	2.45E–06	2.36E–03	2.95E–04
V. cyprinoides	2.6E–06	1.29E–03	3.72E–04

probabilistic risk assessment, the 95th percentile value of HI was very near to the threshold value, similar to the deterministic approach. The TCR of Cd was higher than the threshold value, which implies certain safety risks to humans. For Pb and Cr, the 95th percentile of TCR value was on the edge of the threshold value. The 95th percentile of the risk distribution is employed as a rapid criterion for risk management decisions [35]. Therefore, the presence of heavy metals, particularly Cd, Pb, and Cr, should be seen as a serious warning sign from the perspective of human health. The results of the sensitivity analysis align with findings of [36], both of which highlighted the predominant influence of metal concentration on health risk.

Conclusions

This investigation has added valuable data to the documentation and human health risk assessment of metal contamination in various seafood resources from the Indian coast. Elevated levels of Cu, Zn, Pb, Cd, Mn, and Ni were found in the water, sediment and biota of CES compared to other coastal regions of the world. Cadmium and Pb bioaccumulation raised significant concern by surpassing both international and national food safety standards. The health risk index indicates that consumers are in close proximity to experiencing the synergistic effects of heavy metals, and the most influencing heavy metal during risk analysis was Cd. The carcinogenic risk index indicates that Cd may lead consumers to a chronic cancer risk. Thus, for a better understanding of the scenario, this study recommends a regular monitoring program for all the heavy metals across a broader range of fish species within the study region.

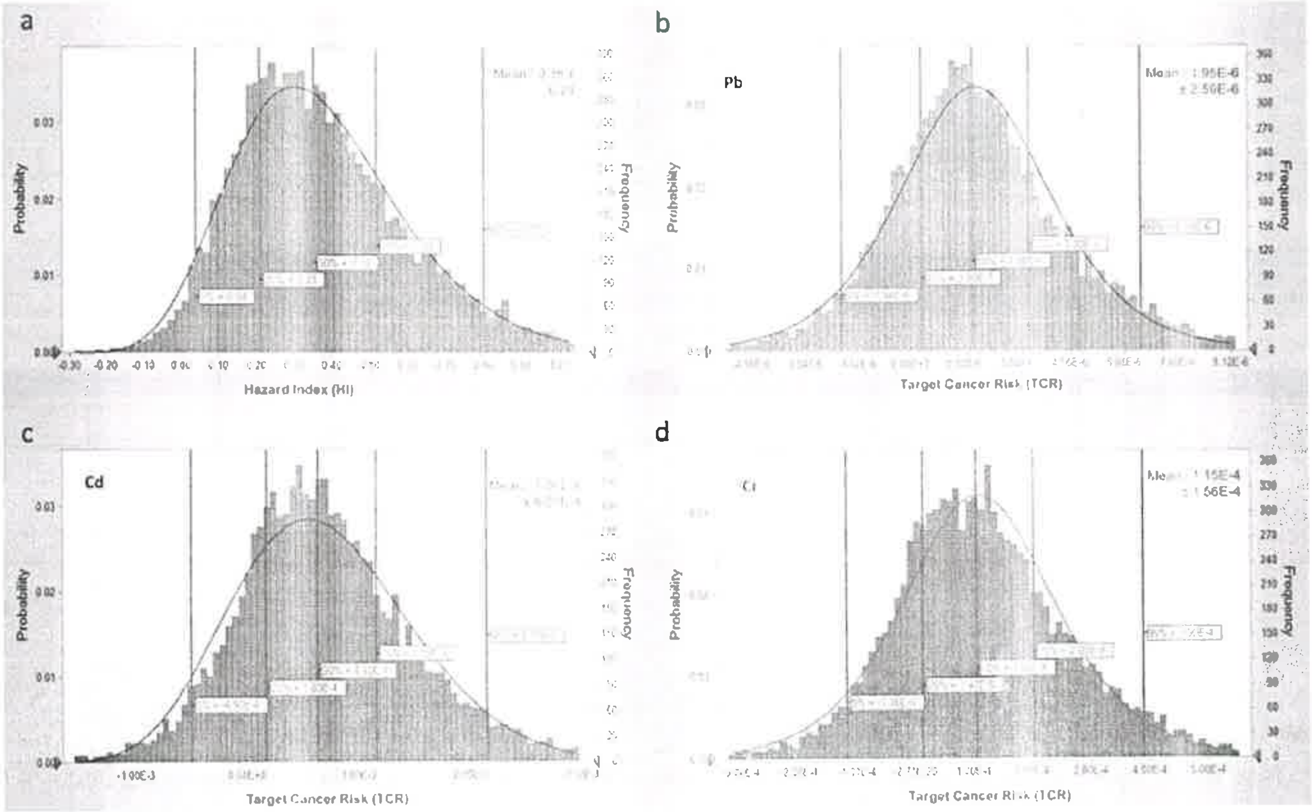

Fig. 5 Simulated values of **a** Hazard index **b** TCR of Pb **c** TCR of Cd **d** TCR of Cr

Materials and methods

Study area

The CES (9° 40′–10° 12′ N and 76° 10′–76° 30′ E) is unique in terms of physiography, climate, geology, hydrology, and other biotic factors [37]. The CES is influenced by heavy rain and freshwater influx during the monsoon. The Southern estuary is influenced by sewage, septage, canal inputs, domestic discharges, fishing and harbor activities, and waste from fish processing units. The Central estuary is a hub for shipping and fishing operations, shipyard activities, dredging activities, oil and ballast water discharge and construction activities. The Central estuary is connected to the Barmouth. The Northern zone is mainly influenced by industrial discharges. Inputs from canals, agricultural fields, hospitals, and municipal discharge also influence this zone [15, 38]. According to the Central Pollution Control Board, the Eloor-Edayar region comes under the 10 highly hazardous waste-contaminated sites in India.

Heavy metal analysis in water and sediment matrix

For heavy metal analysis, water and sediment samples were collected from three distinct zones of the CES, namely the south (S1, S2, S3), central (S4, S5, S6), and northern zones (S7, S8, S9). across the premonsoon (PRM), monsoon (MON), and postmonsoon (POM) periods in 2021 (Fig. 7 and Table S9).To measure the dissolved metals, 500 mL water samples were collected from selected study stations in a sterile polythene bottle and added concentrated nitric acid (HNO_3) to achieve a pH of 2 followed by refrigeration at 4 °C. Subsequent steps included vacuum filtration of the samples through cellulose nitrate membrane filters (0.45 μm) and pre-concentrating them to 25 mL using the ammonium pyrrolidine dithiocarbamate (APDC) and methyl isobutyl ketone (APDC-MIBK) method described by [39]. The analysis for heavy metals was performed using calibrated Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES; Make: Agilent; Model: 5110) with an attached autosampler (Agilent SPS 4) at National Centre for Coastal Research (NCCR), Ministry of Earth Sciences, Govt. of India, Chennai. The instrument was calibrated using known concentrations of metal standards (Merck, Germany).

Sediment samples were collected from selected study stations using a van Veen grab (0.04m^2) and oven-dried at 60 °C. Powdered samples (0.5 g) were subjected to nitric-perchloric acid ($HNO_3/HClO_4$) digestion in block digester (Pelican KES 20L R TS) at an initial temperature of 80 °C for 1 h, 100 °C for 2 h, and 150 °C for 2 h [40]. The resulting

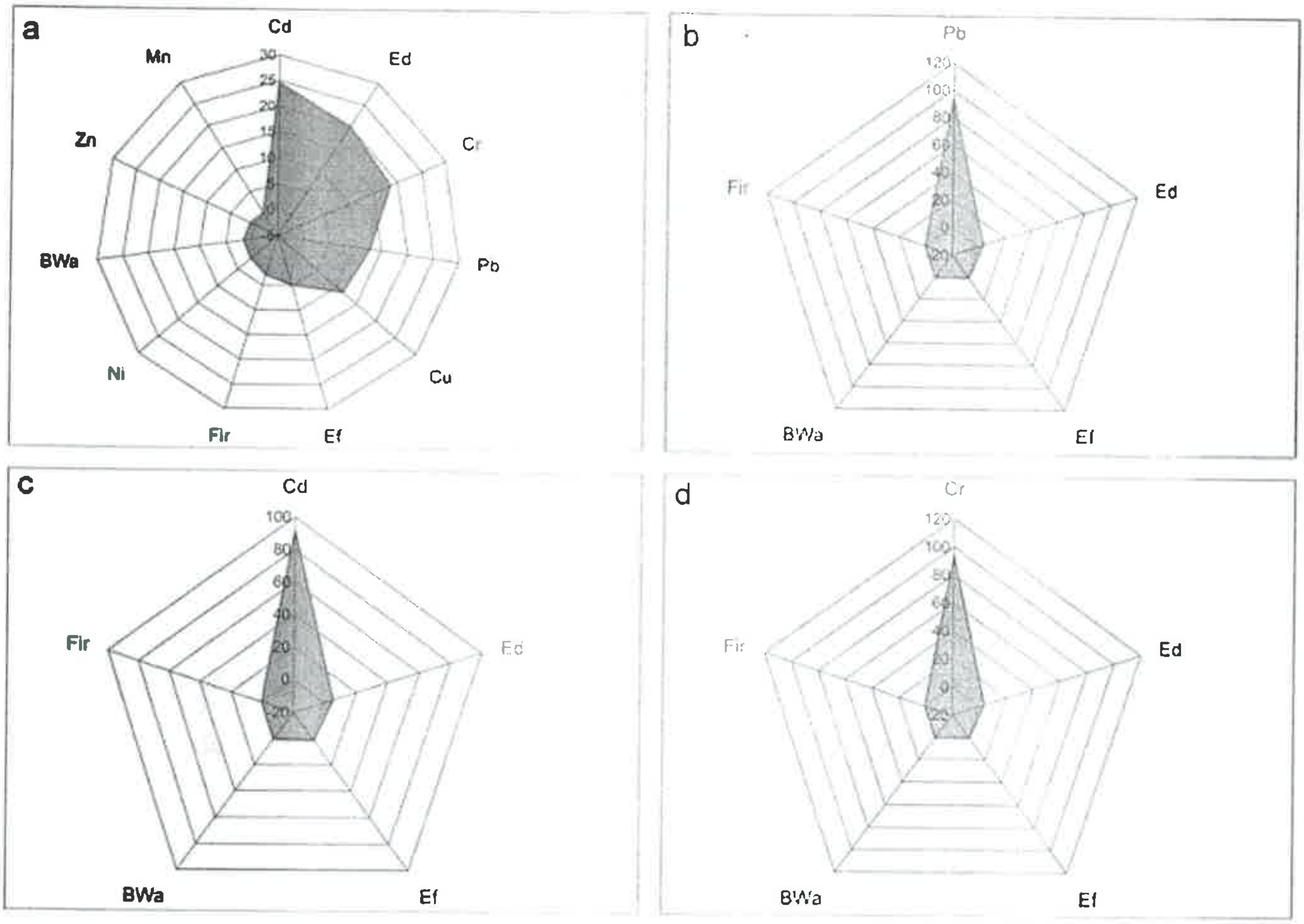

Fig. 6 Sensitivity analysis of input parameters for health risks assessment **a** Hazard index **b** TCR for Pb **c** TCR for Cd **d** TCR for Cr

sample was filtered through a 0.45 μm filter paper and make up it to a volume of 25 ml using Milli-Q water, and metals were analyzed in ICP-OES (Agilent 5110).

Metal pollution indices

Geo-accumulation index (I_{geo}) and contamination factor (CF)

The degree of pollution in sediment was assessed by determining geo-accumulation index (I_{geo}) and contamination factor (CF). The I_{geo}, as outlined by [41], was computed as follows:

$$I_{geo} = \log_2 (C_n)/1.5B_n$$

where

C_n—concentration of metal in sediment, and B_n—geochemical background value in average shale [42]

The determination of the contamination factor was conducted according to the method outlined by [43]

$$CF \equiv \frac{\text{Metal content in sediment}}{\text{Background value of metal}}$$

The classification of I_{geo} and CF is given in Table S1.

Potential ecological risk index (PERI)

The calculation of the potential ecological risk index followed the methodology described by [43]. The risk index (RI) was determined using the following formula:

$$PRFi = TRi \times Cmi/Cbi$$

$$PERI = \Sigma PRFi$$

Here, PRFi represents the potential risk factor associated with a specific pollutant, TRi denotes the toxic response of metals, Cmi signifies the concentration of metal 'i' within

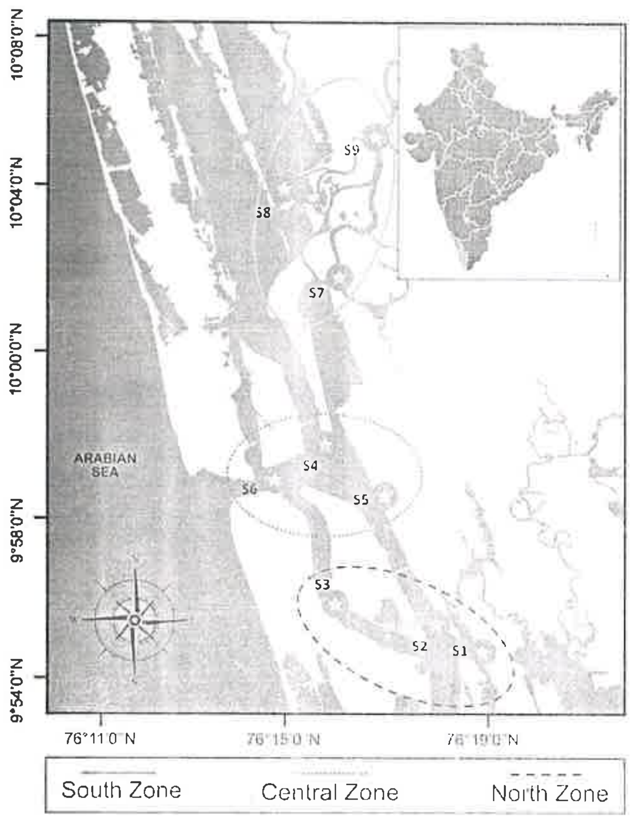

Fig. 7 Map of Cochin Estuarine System showing sampling locations

the sediment, and Cbi refers to the geochemical background value based on average shale data [42]. The classifications for PRFi and PERI are detailed in Table S2.

Sediments quality guidelines (SQGs)

In this study, the concentration of metals was compared to the sediment quality values obtained from the Screening Quick Reference Table (SQUIRT), aiming to assess the toxicity of metals to estuarine biota, as detailed by [44]. Table S3 includes the sediment quality guidelines and terminology utilized in SQUIRT.

Fish sample collection and processing

The study was conducted for a one-year period (2021) on wild populations of fish and shellfish. The samples were collected from the south (S1, S2, S3), central (S4, S5, S6), and northern zone (S7, S8, S9) of the estuary during premonsoon (PRM), monsoon (MON), and postmonsoon (POM) period (Fig. 7 and Table S9). These fish species were selected due to their prevalence in the estuarine environment and their economic significance [45].

Samples were obtained from the fisherman, washed several times with distilled water, placed in an icebox, and sent to the laboratory for further treatment and examination. The collected samples included 10 fish species, 2 crustacean species, and 2 bivalve species (Table S10). Three samples per species were used for the analysis.

The muscle tissue was dried in an oven at 60 °C for 48 h, and powdered using mortar and pestle. The samples were then transferred to boiling tubes and were digested by perchloric acid ($HClO_4$) and nitric acid (HNO_3) in 1:3 ratio at 180 °C in block digester (Pelican KES 20L R TS) until the formation of a clear solution, which then filtered in Whatman filter paper (1 μm) and made up to 25 ml using Milli-Q water [33]. The extracts were analyzed for the metals copper (Cu), zinc (Zn), lead (Pb), cadmium (Cd), manganese (Mn), nickel (Ni), and chromium (Cr) using Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES; Make: Agilent; Model: 5110).

Quality assurance and quality control (QA/QC)

Device validation and calibration were carried out using the fish protein certified reference material (CRM), DORM-4, from the National Research Council of Canada. CRMs underwent acid digestion following the same procedure as tissue samples, with triplicated samples concurrently processed. For sediment, Marine Sediment Certified Reference Material PACS-3 was used as standard. For tissue metal recoveries ranged from 96.4 to 98.23% and for sediment, it was 94% to 100.5% (Table S11). All glassware was pre-cleaned with nitric acid and Milli-Q water, and ultrapure water was used for sample, solution, and standard preparation.

Human health risk assessment

Deterministic risk assessment

Estimated daily intake (EDI)

The EDI values were calculated using equation from [3]:

$$ETDI = \frac{Ed \times Ef \times Fir \times Mc}{BWa \times ATn} \times 10^{-3}$$

Ed is the exposure duration (70 years); Ef the exposure frequency (365 days $year^{-1}$); Fir the fish ingestion rate (53 g $person^{-1}$ day^{-1}); Mc the metal concentration in the muscle tissue of fish; BWa the average body weight (70 kg); and AT the mean exposure period (Ef × Ed).

EDI (μg kg bw^{-1} day^{-1}) values were compared to their respective oral reference doses (RfDs) established by [46]. The RfD values for Cu, Zn, Pb, Cd, Mn, and Ni are 0.04, 0.3, 0.0035, 0.001, 0.14, 0.02, and 0.003 mg kg^{-1} day^{-1}, respectively.

Target hazard quotient (THQ) and hazard index (HI)

THQ and HI values below one (< 1) suggest minimal non-carcinogenic health risks for consumers, while values above one (> 1) indicate the potential health risk [47]. They were calculated using [44]:

$$THQ = \frac{Ed \times Ef \times Fir \times Mc}{BWa \times ATn \times RfD} \times 10^{-3}$$

$$HI = \sum_{n=1}^{n=7} THQ = THQ_{Cu} + THQ_{Zn} + THQ_{Pb} + THQ_{Cd} + THQ_{Mn} + THQ_{Ni} + THQ_{Cr}$$

Target cancer risk (TCR)

The carcinogenic risk, target cancer risk, was calculated using the equation below [48]:

$$TCR = \frac{Ed \times Ef \times Fir \times Mc \times CPS_{o}}{BWa \times ATc} \times 10^{-3}$$

CPS_o—carcinogenic Potency Slope, where 0.5, 6.3, and 0.0085 mg kg^{-1} day^{-1} are for Cr, Cd, and Pb, respectively [46].

The risk of cancer is negligible if the TCR values are less than 10^{-6}, unacceptable if they are greater than 10^{-4}, and acceptable if they are between 10^{-6} and 10^{-4} [49].

Probabilistic risk assessment and sensitivity analysis

Monte Carlo simulation (MCS) was used in Oracle Crystal Ball v11.2 software to address uncertainties in health risk assessments with 10,000 random simulations [50]. A sensitivity analysis was also performed in the software to determine the impact of input parameters on risk assessment (v11.2).

Data analyses

One-way analysis of variance (ANOVA), followed by the Tukey post hoc test, was used to detect spatiotemporal differences across water, sediment, and biota. Pearson correlation tests were employed to assess the relationships between specimen length, weight, and heavy metal concentrations. Both ANOVA and correlation analyses were performed using IBM SPSS Version 25 software. Monte Carlo simulation and sensitivity analysis were conducted using Oracle Crystal Ball v11.2 software. A shade plot was generated using PRIMER v7 (Plymouth Routines in Multivariate Ecological Research) statistical software to pinpoint the metal contributing to variances among study stations. Origin Pro 2022 was employed to create all graphical representations. Each measurement was repeated three times, and the results were presented as mean ± standard deviation.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s13530-024-00212-0.

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Declarations

Conflict of interest Kariyil Veettil Neethu, Punnakkal Hari Praved, Neethu Xavier, Naduvath Deepak Sankar, Hanse Antony, Sivasankaran Bijoy Nandan, Panneerselvam Karthikeyan, Shambanagouda Rudragouda Marigoudar, and Krishna Venkatarama Sharma declare that they have no conflicts of interest.

Ethical approval This article contains no studies with human participants or animals performed by any authors.

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BEFORE THE HON'BLE NATIONAL GREEN TRIBUNAL SOUTHERN BENCH

AT CHENNAI

O.A. 246 OF 2024

In the matter of News item title "Zinc, Lead…fish in Kochi estuary are heavy, daily intake risky" appeared in The Times of India dated 06.05.2024

…Petitioner

Vs.

National Centre for Coastal Research, through its Director,Chennai & Ors.,

…Respondents

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S. JANARTHANAM(877/91)
COUNSEL FOR 1ST RESPONDENT